Air Mass Characterisation During EOPACE: Aerosol Composition and Concentration

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LONG-TERM GOALS

The overall aim of this project is to characterise the aerosol physico-chemical properties for air masses found within the littoral zone for a wide range of conditions in order that their impact upon the propagation of visible and infra-red radiation may be quantified. The aerosol particles found within these air masses have a variety of sources, both natural and anthropogenic and are usually dominated by the accumulation mode (particle radii from about 0.05 to 0.5μm). To this prevailing aerosol are added particles of more local origin which are generally larger, including surf-generated sea spray droplets which form the subject of companion investigations.

OBJECTIVES

The primary objective of the current work has been the characterisation of accumulation mode aerosol ($0.05 < r < 1.5$ μm), within the littoral zone at a variety of locations. In order to achieve this objective, a thermal analytical, 'volatility' technique has been utilised which is based on an Optical Particle Counter (OPC). Simultaneous soot carbon loadings have been recorded using an aethalometer in order to validate the volatility measurements and simultaneously provide a useful indication of the magnitude of anthropogenic aerosol input into the measured air masses.

During the EOPACE field campaign at the Army Research Facility, Duck, North Carolina, a Tandem Volatility Differential Mobility Analyser (TVDMA) was operated by Dr Barbara Brooks alongside the equipment deployed for all the other EOPACE campaigns. The TVDMA was developed for use in ground-based and airborne projects to extend the volatility analysis to smaller particles (down to about 5nm radius) as well as to overcome some of the deficiencies of the OPC approach, generally associated with more complex aerosol mixtures.

APPROACH

OPC Volatility

The original instrumentation (O'Dowd *et al*, 1992, O'Dowd & Smith, 1993, Smith & O'Dowd, 1996) consisted of a Particle Measuring Systems Active Scattering Aerosol Spectrometer Probe (ASASPX) optical particle counter, radius range 0.05 to 1.5μm, preceded by a quartz tube wrapped in a heater coil.

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Volatility analysis was performed by rapidly heating the quartz tube to 1000 °C and then cooling it passively to ambient; cycle time being approximately 1 hour, while constantly monitoring the output of the ASASPX. The OPC-based instrument has been operated throughout the EOPACE campaigns with a notable degree of success, especially as changes in aerosol properties during these projects were generally very gradual and the hourly operating cycle was not a major impediment. However, changes in spectral shapes upon heating of the aerosol under such conditions could not always be unambiguously ascribed to either a shift to smaller sizes associated with the volatilisation of material from internally mixed particles, or to a general decline in particle numbers resulting from the loss of the more volatile members of an externally mixed aerosol. Also, the heater system operation meant that the least time was spent at the highest temperatures where resultant particle concentrations were inevitably at a minimum which resulted in poor sampling statistics for the more least volatile particles. A further problem, of greater consequence for the EOPACE project, sometimes arises with sea salt aerosol in which some aerosol particles larger than the upper size limit of the OPC shatter within the heater tube, giving an apparent increase in particle numbers with heating. The TVDMA instrument overcomes these problems by employing an electrostatic classifier to select specific particle sizes prior to their examination by a heater system coupled to a scanning differential mobility analyser.

DMA Volatility

The TVDMA selects a monodisperse dry aerosol population for analysis from the poly-disperse sample stream using a TSI 3071 Electrostatic Classifier. The classifier voltage is chosen to correspond to a specific radius, the centre/filter radius, with the spread in the size distribution being dependent upon the ratio of the mono-disperse to sheath flows. Filter radii of 30nm, 50nm, and 70nm with a distribution width of ±10 % were used throughout.

The volatility analysis is performed by continually monitoring the size distribution of the filtered aerosol as the temperature is varied. The monitoring system consists of a TSI 3071 Electrostatic Classifier coupled with a TSI 3010 Condensation Particle Counter (CPC) to form a Scanning Mobility Particle Counter (SMPS). The SMPS generates a 64-channel distribution for sizes 5 to ~200nm in 2 minutes. The mono-disperse flow through the tandem classifier system is controlled by the 1 l/min critical orifice in the CPC, while the excess flow of both classifiers is controlled by 10 l/min critical orifi (Jokinen & Makela, 1997; Landau & Lifshitz, 1987). Sheath flows in both classifiers were adjusted manually to match the excess flows.

As mentioned above the volatility heater systems based on a heating element wrapped around a quartz glass tube proved to be both slow in response and to have poor temperature stability. The heater developed for this project consisted of a ¼ inch diameter stainless steel tube brazed around 20cm of which was a 500W heating element. The element comprised a thin walled stainless steel tube containing the electrical element embedded in an electrically insulating/thermally conducting ceramic. The temperature at the centre of the heater tube could be varied from ambient up to 1000ºC with an accuracy of ±10ºC and a considerable improvement in both response time and stability from the previous configuration was achieved. This system was developed by Mr Martin Hill and employed forced cooling under microprocessor control, allowing for user-defined temperature sequences.

WORK COMPLETED

Extensive aerosol measurements have been made at the seaward end of the USACE Field Research Facility pier during the EOPACE IOP#9 field project at Duck, NC (February - March 1999). The thermal analytical system was used to record volatility aerosol composition data. Soot carbon loadings were measured using an aethalometer and aerosol measurements using an FSSP-100 and an OAP-230X were made throughout the project. Analysis and validation of these data and those from all the earlier EOPACE campaigns has been completed and work on various publications is proceeding in collaboration with other EOPACE participants. In addition to this work, results from the TVDMA volatility system deployed for the first time at Duck have been analysed and compared with results from the OPC system. During this field campaign, these instruments were mounted close to the end of the pier at the Duck facility alongside the other instrumentation deployed by this group.

RESULTS

Although the two instruments have very different measurement ranges, for a 70nm filter size, the resultant spectrum from the DMA based unit should overlap with the bottom end of the OPC spectrum. This overlap was checked by setting the temperature of both heaters to 30 ºC and the resultant spectra were compared to confirm that no unanticipated distortions in the spectra occurred.

Figure 1 shows the temperature dependence of two particle radii, 30 and 50nm, throughout a large portion of the Duck field measurements, during which the temperature range of the TVDMA ranged from 30 to 900ºC. In the case of the 30nm filter size, it may be noted that the majority of the aerosol was volatilised by about 200ºC, indicating the predominance of ammonium sulphate in these smaller aerosol particles. Occasionally, material volatile at lower temperatures is seen, which indicates the

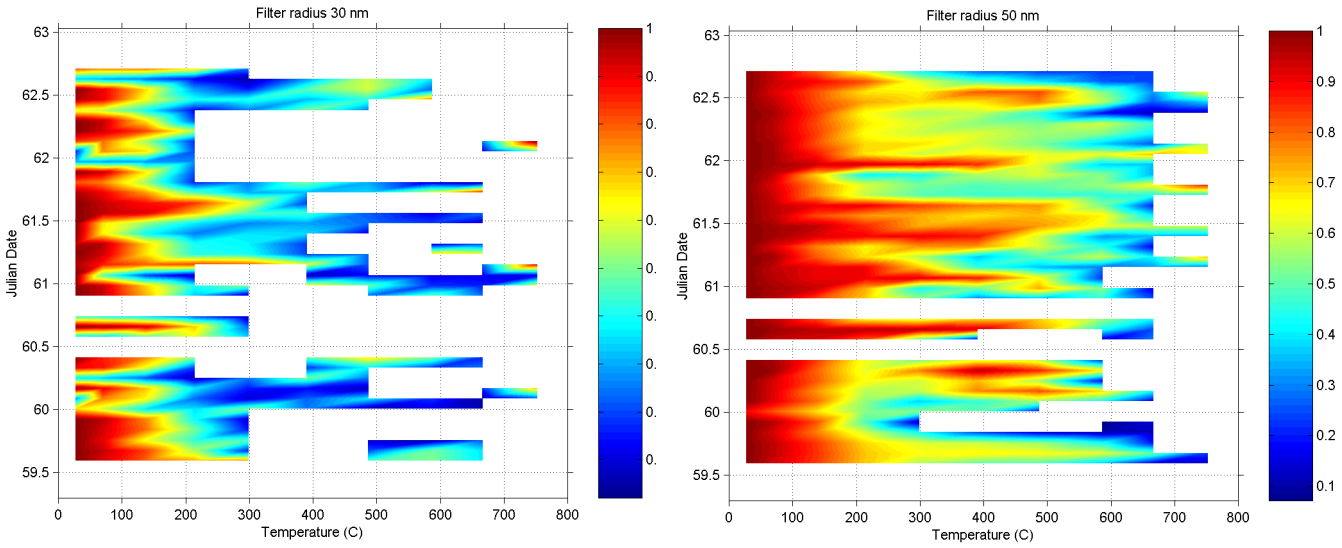

Figure 1: Behaviour of the normalised mode radius (as a fraction of the filter radius) with temperature for 30nm and 50nm filter radii, as measured by the TVDMA

presence of sulphuric acid or even organic components, while these is an indication of less volatile material toward the end of Julian Day 61. The 50nm aerosol is clearly less volatile for extended

periods throughout the Duck project indicating the presence of sea salt (volatile above about 500 ºC). From the behaviour observed in the laboratory it could be inferred that the aerosols are internally-mixed sea salt/ammonium sulphate composites. It was observed that sea salt volatilised by 700ºC in both systems while ammonium sulphate volatilised at 250ºC. While these results are similar to those found with the older OPC system, the DMA observations show more clearly the dependence of the particle composition on size.

On occasions during the Duck field trial, the DMA was operated without the heater system in order to provide higher time resolution information on the physical aerosol properties. One such sequence is shown in Figure 2 and illustrates the dominance of the accumulation mode aerosol (radii around 0.1µm) through much of the middle period, with the aerosol moving toward smaller sizes at the end of the sequence and evidence of a smaller mode around 20nm radius at the beginning of this period. During this time sequence, the meteorological records showing a shift in wind direction from 100º to 360º and humidity changes from values of about 90% to 50%. Carbon loadings also increased substantially over the period and, thus, the changes in aerosol physico-chemistry are consistent with a shift from a maritime aerosol to a much drier one with strong influences from the urban areas some distance to the north of the field site.

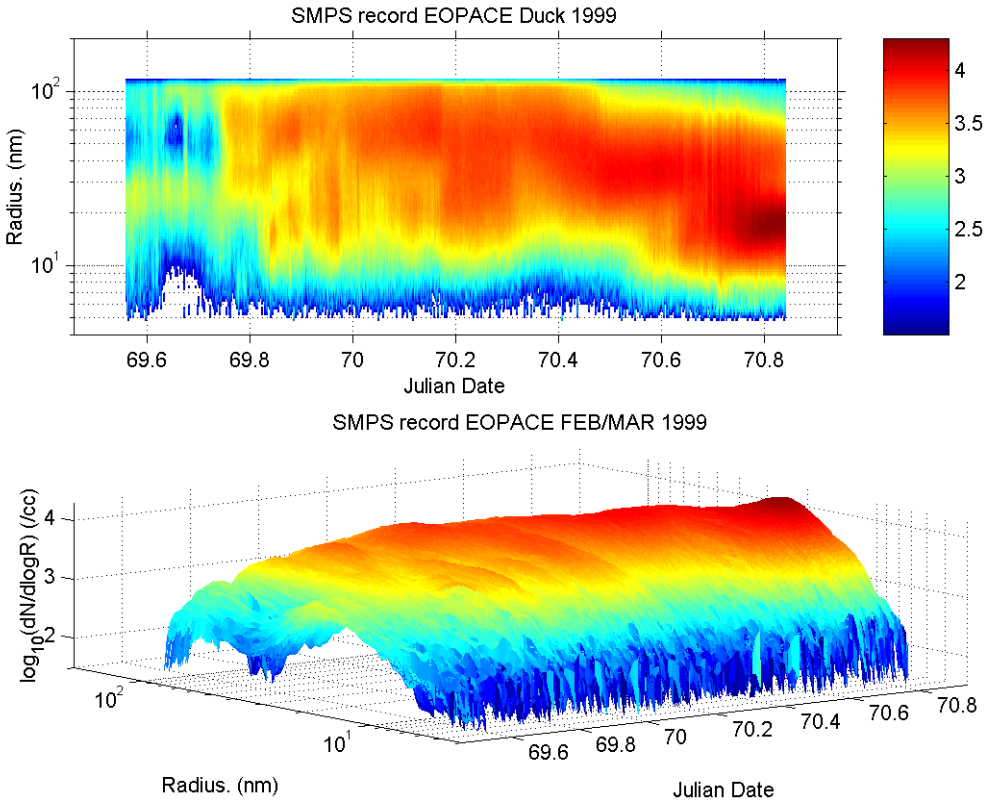

Figure 2: Temporal sequence showing two views of the smaller aerosol particle spectrum during the Duck field campaign.

IMPACT/APPLICATION

These measurements are useful in defining the variability in aerosol concentrations and composition found within the littoral zone and attempt to identify meteorological and oceanographic parameters which are more easily obtained than direct aerosol measurement which may be used to estimate air mass characteristics. The generally low loss rates of the accumulation mode particles which dominate air mass characteristics results in their conservation within a given air mass (with allowances for changes in relative humidity). Hence, current mesoscale meteorological models should be capable of development to incorporate aerosol sources, sinks and transport processes in order to provide predictions of these air mass characteristics. These measurements provide a topographic map of aerosol for use in such mesoscale models.

TRANSITION

This investigation is a component of the EOPACE programme whose purpose is to characterise the coastal atmospheric environment and determine the impact of surf-generated aerosols on atmospheric extinction within the coastal environment for evaluation of electro-optical systems performance.

RELATED PROJECTS

The UK Ministry of Defence provided funding via DERA Portsdown Contract No: SSDH300037 to cover participation in the surf zone studies which form an associated element of the EOPACE programme. DERA have also provided funding under Contract No: SSDW3/0049 to continue with the analysis of the surf zone aerosol production analysis.

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